LA-UR -93- 2901

Mechanical Design Parameters for Detection of Nuclear Signals by Magnetic Resonance Force Microscopy

Gregory J. Moore, Jack A. Hanlon, Bruce Lamartine, Marilyn Hawley, Johndale C. Solem, Sidney Singer, John J. Jarmer, Seppo Penttila and Laurel O. Sillerud

11th International UACEM Symposium Proceedings

Los Alamos National Laboratory
Los Alamos New Mexico 87545

11th International UACEM Symposium Proceedings, Boston, MA, August 18-21, 1993

MECHANICAL DESIGN PARAMETERS FOR DETECTION OF NUCLEAR SIGNALS BY MAGNETIC RESONANCE FORCE MICROSCOPY

Gregory J. Moore, Jack A. Hanlon, Bruce Lamartine, Marilyn Hawley, Johndale C. Solem, Sidney Singer, John J. Jarmer, Seppo Penttila and Laurel O. Sillerud

Biomedical NMR Facility
Division of Life Sciences
Los Alamos National Laboratory,
Los Alamos, New Mexico 87545
U.S.A.

Ryszard J. Pryputniewicz

Center for Holographic and Laser Technology
Department of Mechanical Engineering
Worcester Polytechnic Institute
Worcester, Massachusetts 01609
U.S.A.

ABSTRACT

Recent theoretical work has shown that mechanical detection of magnetic resonance from a single nuclear spin is in principle possible. This theory has recently been experimentally validated by the mechanical detection of electron spin resonance signals using microscale cantilevers. Currently we are extending this technology in an attempt to detect nuclear signals which are three orders of magnitude lower in intensity than electron signals. In order to achieve the needed thousand-fold improvement in sensitivity we have undertaken the development of optimized mechanical cantilevers and highly polarized samples.

Finite element modeling is used as a tool to simulate cantilever beam dynamics and to optimize the mechanical properties including Q, resonant frequency, amplitude of vibration and spring constant. Simulations are compared to experiments using heterodyne hologram interferometry. Nanofabrication of optimized cantilevers via ion milling will be directed by the outcome of these simulations and experiments.

Highly polarized samples are developed using a three-fold approach: 1) high magnetic field strength (2.5T), 2) low temperature (1K), and 3) use of samples polarized by dynamic nuclear polarization. Our recent experiments have demonstrated nuclear polarizations in excess of 50% in molecules of toluene.

1. INTRODUCTION

Magnetic resonance force microscopy (MRFM) is the use of mechanically detected magnetic resonance (MR) signals to image small objects with high resolution and to simultaneously identify the chemical species. Until recently, MR signals have only been detected by electrical means which have a sensitivity limit of approximately 10^{15} nucleons. Recent work by Sidles et al has shown that the theoretical limit for mechanical detection is a single nucleon[1-4]. Rugar et al have experimentally validated this theory by the mechanical detection of electron spin resonance (ESR) signals using microscale cantilevers[5,6].

Currently, we are extending this technology in an attempt to detect nuclear signals which are three orders of magnitude lower in intensity than electron signals. A subsequent goal is to determine the ultimate experimental sensitivity of the technology with the goal of detecting a single nuclear spin. The main focus of our investigations to date has been to improve the sensitivity, using both simulation and experiment to design and develop optimized mechanical cantilevers and highly polarized samples.

2. BACKGROUND

A schematic diagram of the NMR force microscope probehead is shown in Fig. 1. The probehead is an evacuated chamber and consists of a mechanical cantilever, similar to those used in atomic force microscopy, to which the sample is mounted. The parameters of typical commercial cantilevers include a Q of 2000 and a resonant frequency of 8 KHz. A small magnetic sphere provides the magnetic field gradient which is responsible for the high resolution of the microscope. An rf coil allows for excitation of the sample while a B_0 modulation coil provides the capability to slowly sweep the main magnetic field. When the sample is at a critical distance with respect to the field generated by the magnetic sphere, the cantilever will oscillate at its resonant frequency. The cantilever vibrations can be detected by the fiber optic interferometer which has a sensitivity on the order of 0.1 Angstroms[7]. Positioning of the components is accomplished with various PZT translation devices. This probehead is designed to be inserted into a He^3-He^4 dilution refrigerator which can maintain a temperature of 10mK. The dilution refrigerator apparatus is contained within a 2.5T superconducting vertical bore magnet. A microwave wave guide is attached to the probehead to allow for dynamic nuclear polarization[8] (DNP) of the sample. At 2.5T, 70 GHz microwaves are used to irradiate the sample for the DNP procedure.

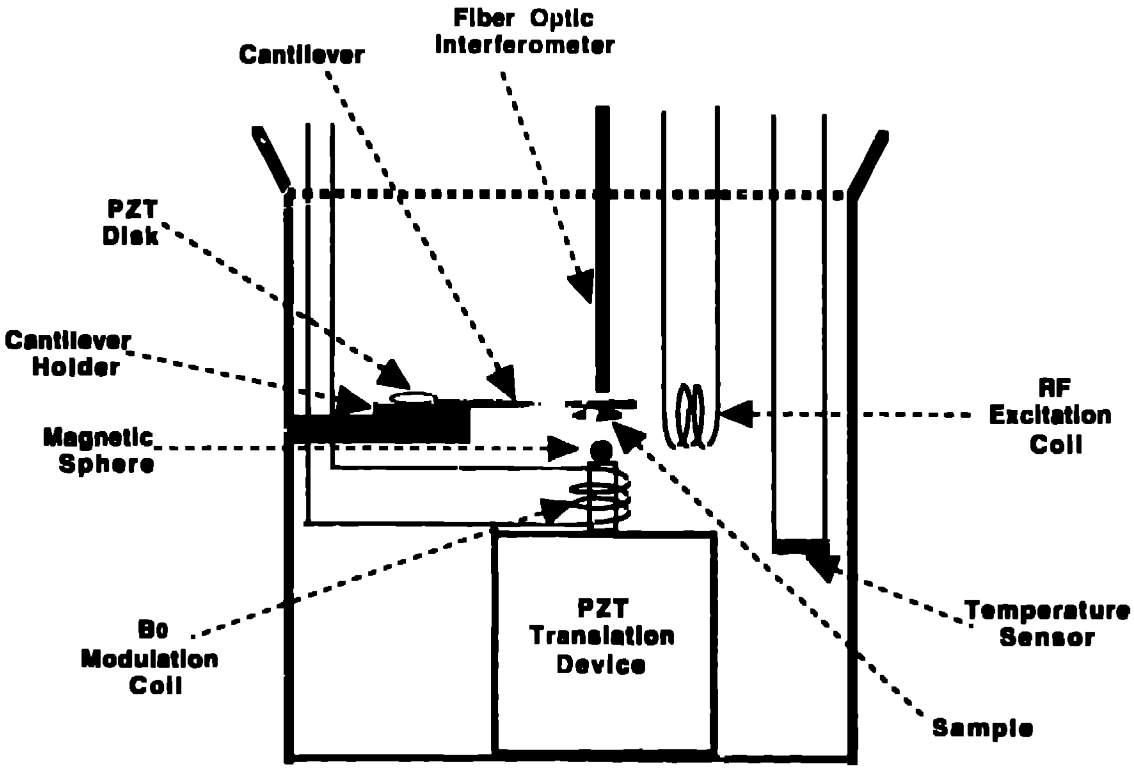

Fig. 1 NMR force microscope probehead.

The sensitivity of the NMR force microscope is determined by Eq. 1

$$\mu_{min} = (2\,k\,k_B\,T\,\Delta\nu / Q\,\omega_c)^{1/2} / |\nabla B| \qquad (1)$$

where μ_{min} is the minimum magnetic moment detectable in a sample, k is the spring constant, k_B is Boltzman's constant, T is temperature, $\Delta\nu$ is the detection bandwidth, Q is the quality factor, ω_c is the resonance frequency of the cantilever and ∇B is the magnetic field gradient. In order to improve the sensitivity of the NMR force microscope, each of the parameters in Eq. 1 should be optimized.

Table 1 shows the parameters for Eq. 1 at their current status with present day technology and also where they need to be in the future to achieve single nucleon sensitivity. Plugging in the current status parameters into Eq 1 yields a minimum magnetic moment sensitivity of 1.5×10^{-17} J/T. Since a single proton magnetic moment is 1.4×10^{-26} J/T, an improvement of greater than 10 orders of magnitude in sensitivity is needed. Plugging in the parameters in the goal column gives a sensitivity of 2.2×10^{-28} J/T, more than enough to see a single proton. The parameters in the goal column were carefully chosen such that they should be achievable with careful design and fabrication using currently available materials and technology.

Table 1 Technical Challenges to Achieve Single Proton Sensitivity.

<table>
<tr><th>Parameter</th><th>Current Status</th><th>Goal</th><th>Sensitivity Increase</th></tr>
<tr><td>Cantilever Length</td><td>200 μm</td><td>5 μm</td><td rowspan="2">$3X10^1$</td></tr>
<tr><td>Cantilever Frequency</td><td>8 KHz</td><td>7.2 MHz</td></tr>
<tr><td>Cantilever Q</td><td>2000</td><td>$2X10^6$</td><td>$3.1X10^1$</td></tr>
<tr><td>Cantilever Spring Constant (k)</td><td>0.1 N/m</td><td>0.01 N/m</td><td>3.1</td></tr>
<tr><td>Magnetic Field Gradient</td><td>60 T/m</td><td>$8x10^7$ T/m</td><td rowspan="2">$1.3X10^6$</td></tr>
<tr><td>Magnetic Sphere Diameter</td><td>1 mm</td><td>300 Å</td></tr>
<tr><td>Temperature</td><td>300 K</td><td>1 K</td><td>$1.7X10^1$</td></tr>
</table>

3. EXPERIMENTAL APPROACH

As discussed above, we are extending current MRFM technology in an attempt to detect nuclear signals and also to explore the ultimate achievable sensitivity of the technology. This work is proceeding in two areas: 1) the simulation and design of cantilevers and 2) the development of polarized samples. Optimal mechanical cantilevers provide a more sensitive detection scheme, while highly polarized samples give an increased signal intensity.

To date, commercially available cantilevers designed for use in scanning tunneling (STM) and atomic force microscopy (AFM) have been used in MRFM instruments. However, the properties of these cantilevers are not optimized for MRFM experiments. Our approach has been to use finite element modeling (FEM) as a tool to simulate cantilever beam dynamics and to optimize the mechanical properties including Q, resonant frequency, amplitude of vibration and spring constant. Simulations are then compared to experiments using heterodyne hologram interferometry (HHI).

Previous, MRFM experiments have been performed at room temperature and at low magnetic field strengths resulting in polarizations on the order of 10^{-6}. The nuclear polarization in a nuclear magnetic resonance (NMR) experiment is described by Eq. 2,

$$(n\uparrow - n\downarrow) / n \quad \gamma \hbar B_0 / 2 k_B T \tag{2}$$

where $(n\uparrow - n\downarrow) / n$ is the nuclear polarization, γ is the gyromagnetic ratio, $\hbar$ is Plank's constant, B_0 is the magnetic field strength , k_B is Boltzman's constant and T is the temperature. Our focus has been to improve the achievable nuclear polarizations using a three-fold approach: 1) increased magnetic field strength (2.5T), 2) decreased temperature (1K), and 3) use of samples polarized by dynamic nuclear polarization[8].

4. RESULTS

An example of FEM and HHI of a cantilever beam is shown in Fig. 2. This cantilever beam is in second torsion mode at approximately 5.2KHz. Note the excellent agreement between the beam dynamics predicted by simulation and those found experimentally. Nanofabrication of optimized cantilevers will ultimately be directed by the outcome of these simulations and experiments.

A particularly promising cantilever design which we are currently investigating in detail is diagrammed in Fig. 3. Single crystal quartz will be used as a material because it is well characterized and can have Q values in excess of 10^6. The small size of the beam combined with its aspect ratio and the reduced mass due to the holes should provide a high resonant frequency. The sphere and sample are interchanged from those shown in Fig. 1 for ease of operation.

An example of proton NMR detection of a sample of toluene polarized by DNP is shown in Fig. 4. The net polarization achieved in this case was 56.5%. The dashed line represents the polarization observed without the DNP procedure, which under these conditions (2.5T and 1K) is 0.25%.

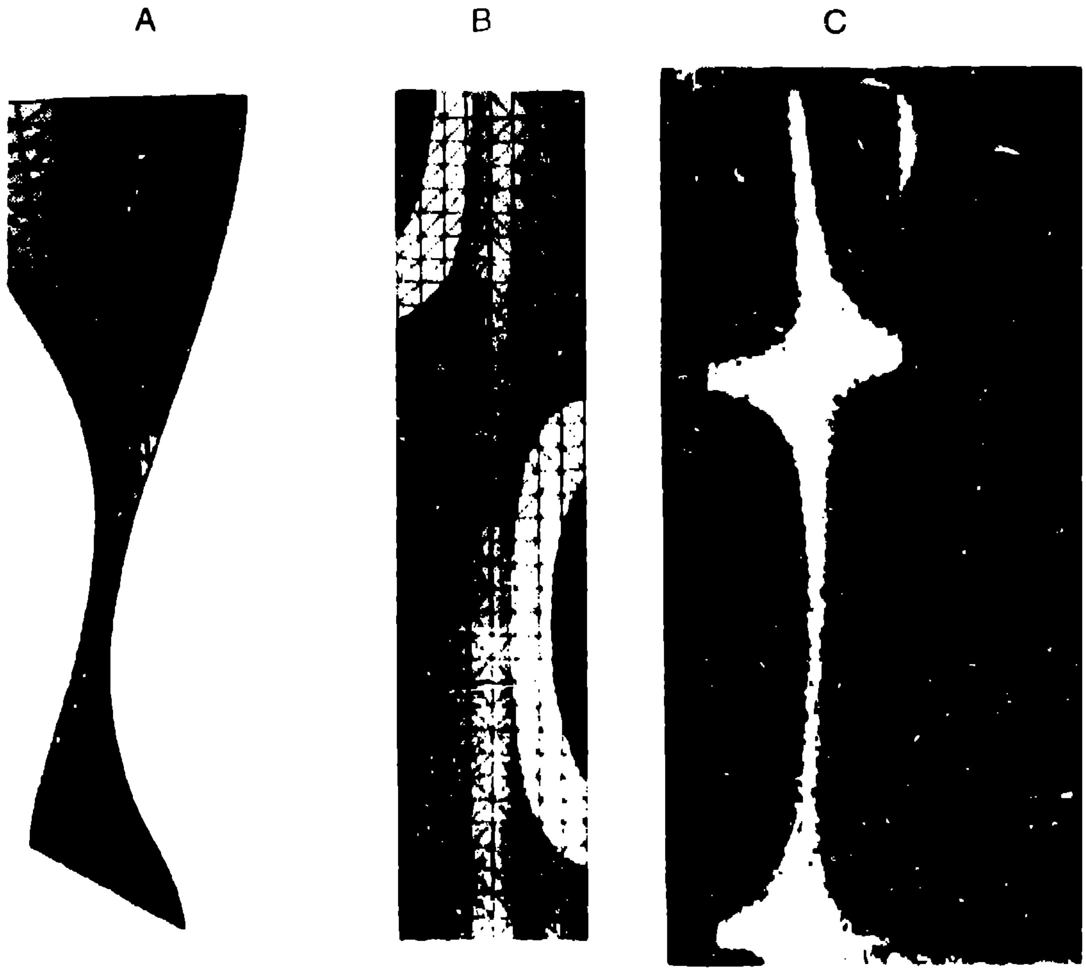

Fig. 2 Cantilever beam in second torsion mode. (A and B) Finite element modeling of cantilever beam dynamics; side and top views are shown, respectively. (C) Heterodyne hologram interferometry of cantilever beam resonating at 5.2KHz.

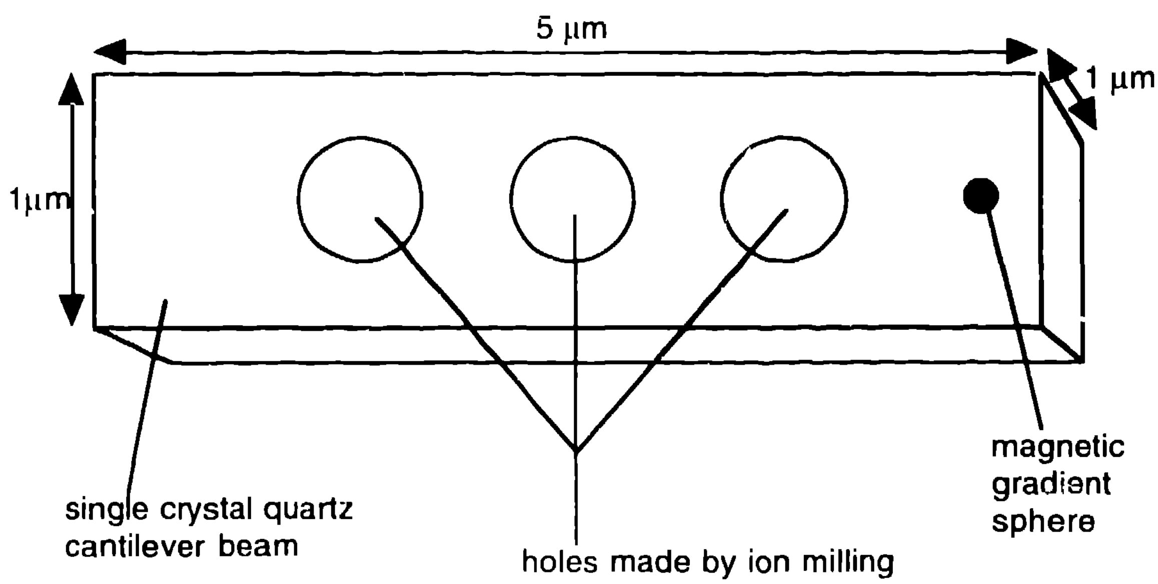

Fig. 3 Proposed cantilever beam design for increased sensitivity.

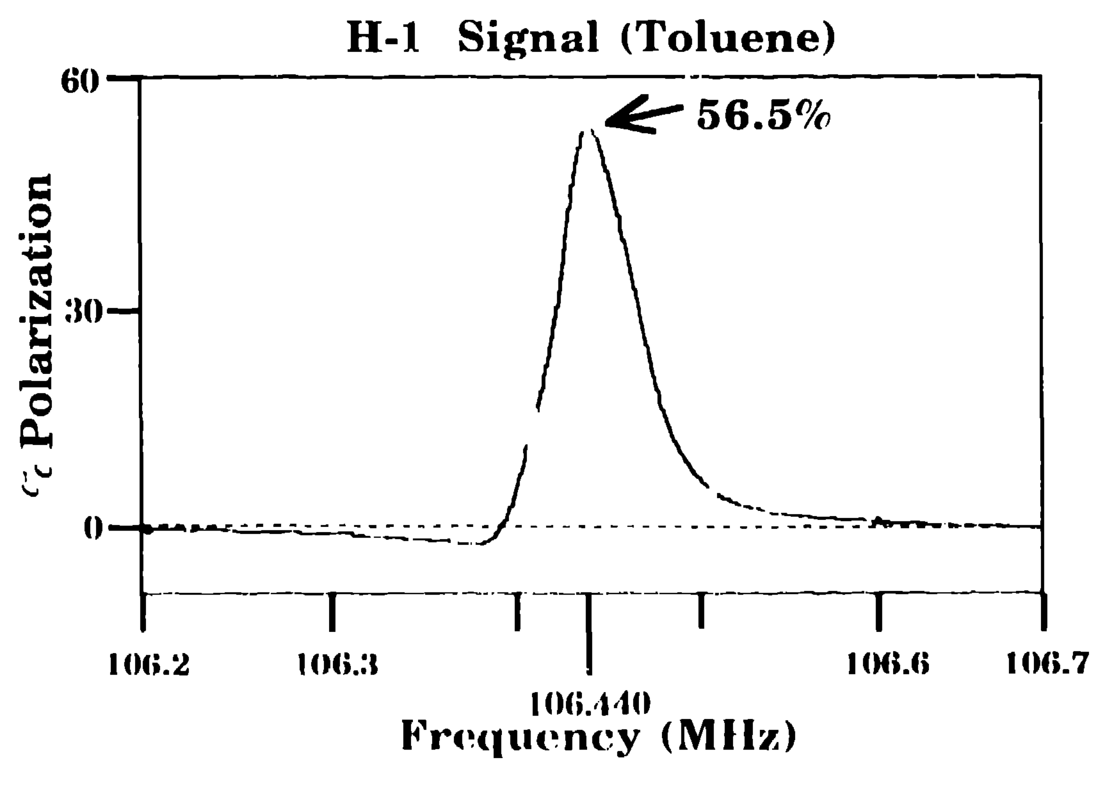

Fig. 4 Example of nuclear polarization by DNP in a sample of Toluene at 1K and 2.5T. Dashed line represents nuclear polarization without DNP.

5. DISCUSSION

The large improvement in nuclear polarization achieved by the DNP procedure should dramatically increase the signal-to-noise of the NMR force microscopy experiment. Using this added signal intensity, a straightforward calculation of the sensitivity of this experiment based on previous work[5] shows that we should be able to design an experimental device capable of detecting on the order of 10^9 nuclear spins. Further improvements in sensitivity should be achieved by the design and fabrication of more sensitive cantilevers with large Q and higher resonant frequencies based on the outcome of simulation and experiment.

Overcoming the technical challenges facing the development of a single molecule NMR force microscope will require considerable effort. However the potential advantages of such a tool over present day technology such as scanning tunneling and atomic force microscopy, justify this effort, namely:

- The imaging is noncontact and nondestructive.
- The imaging field is three dimensional and reaches below the scanned surface.
- NMR is the only method which gives direct chemical information about the sample.

The successful development of an NMR force microscope would make direct molecular imaging a reality and would provide a valuable tool for diagnostic and structural studies in materials science, microelectronics, molecular biology and pharmaceutical development.

6. ACKNOWLEDGMENTS

The authors would like to acknowledge helpful discussions with J.A. Sidles of the University of Washington and D. Rugar of IBM during the early stages of this research. This work was performed under the auspices of the U.S. Department of Energy.

7. REFERENCES

1. J.A. Sidles, Noninductive Detection of Single-Proton Magnetic Resonance, *Applied Physics Letters* **58**(24), 2854, 1991.

2. J.A. Sidles, Folded Stern-Gerlach Experiment as a Means of Detecting Magnetic Resonance in Individual Nuclei, *Phys. Rev. Lett.* **68**, 1124, 1992.

3. J.A. Sidles, J.L. Garbini, and G.P. Drobny, The Theory of Oscillator-Coupled Magnetic Resonance with Potential Applications to Molecular Imaging, *Rev. Sci. Instr.* **63**, 3881-3899, 1992.

4. J.A. Sidles and D. Rugar, Signal-to-Noise Ratios in Electrical and Mechanical Detection of Magnetic Resonance, submitted to *Phys. Rev. Lett.*

5. D. Rugar, C.S. Yannoni, and J.A. Sidles, Force Detection of Magnetic Resonance, *Nature* **360**,563, 1992.

6. O. Zuger and D. Rugar, Magnetic Resonance Force Microscopy, submitted to *Science*.

7. T.R. Albrecht, P. Grutter, D. Rugar, and D.P.E. Smith, A Low Temperature Fiber Optic Interferometer, *Ultramicroscopy* **42-44**, 1638, 1992.

8. C.D. Jefferies, *Dynamic Nuclear Orientation*, Interscience, New York, 1953.